COMPOSITE MATERIAL BEHAVIOR UNDER HIGH ELECTRIC FIELDS:
II- MATERIAL SWITCHES TO " 'HIGH CURRENT CARRYING' " STATE.

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Summary:

Electrically conductive silver filled epoxy EC F-563 preform exhibit intermittent switching to 'high current carrying' state under applied high electric field (this phenomena occurs in the same materials which were undergone switching to 'high resistive' state at lower applied voltages). This behavior has been reported in the literature before for a wide variety of composites of metal particles in a background of insulating media and metal-insulator-metal junctions. The mechanism of "forming" is named responsible for such a behavior. We believe that this switching is an intrinsic property of metal-insulator composites. An appropriate microscopic picture of these compounds will be discussed together with influence of boundaries and 'the complications caused by presence of high electric field coupled with space charge. The industrial applications of a material with a well-defined sharp threshold switching voltage are numerous.

Key **Words-** intermittent switching, hybrid circuit reliability, silver filled conducting epoxy preform, tunneling, fluctuation, trap centers, defects, surge arrest materials.

Reader Aids-

Purpose: To describe observation of intermittent switching to 'high current carrying' state in silver filled conductive epoxy preforms.

Results Useful To: Reliability Engineers concerned with die attach/hybrid circuits electrical intermittancy problems.

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INTRODUCTION:

In the first section of this paper [1], we described the observation of intermittent switching to a 'high resistive' state in silver filled epoxy preform Ablefilm ECF-563 (Ablebond Co.) under applied high electric field. We argued that this behavior is an intrinsic property of a composite of metal particles in a background of insulating matrix. We further brought the attention of our readers to the presence of defects in the insulator surrounding the ndividual metal particles. Strongly localized defects can represent space charges which can generate high electric fields in a direction opposing the current and capable of stopping it. We described further that bulk of EC F-563 composite structure is covered with a thin layer of epoxy matrix on the surface. Injection and ejection of current between the contact pads and the bulk of epoxy is also subject to the presence of defects. for a 0.003" thick sample of ECF-563 sandwiched between 2 gold contacts, a threshold voltage of 0.4-1.9 V existed for switching to a 'high resistive' state. A device based on this property custom designed to a fixed threshold voltage can be used in wide applications such as surge arrest materials or for ESD protection of sensitive electronics instruments.

In continuation of the experiments reported n [1], we have performed additional tests which will be discussed next

EXPERIMENT:

ECF-563 preform sheet of 0.003" thickness was cut in a form of a strip and laid down flat over two adjacent 50 ohm gold microstriplines. These lines were fabricated on a Al_2O_3 substrate soldered on a copper block within a fixture. The width of gold lines were 0.023" and the width of EC F-563 strip was ~ 0.050". The sample of ECF-563 epoxy was cured at 125o C for 2 hours. The fixture was made such that two coaxial connectors can be mounted on the side walls with their center pins protruding through the wall and soldered to the gold microstriplines. Two-probe resistivity measurement of the sample was done with Keithley 237 instrument. Due to the specific geometry described, current path is lateral within the bulk of strip (-0.5" long) but perpendicular to the surface of epoxy at the contacts. As the voltage increased, the I-V characteristic of the sample starts to become nonlinear approximately at 60 V when its current quickly reached 100 mA (our measurement limit). Figure 1 depicts the results of this measurement in a log-log scale.

we have also looked at a similar sample under Tektronix 576 curve-tracer (narrow-pulse operation mode) and a similar behavior was obtained. The 2-probe resistance of the sample initially was $>100\,K\Omega$. Nonlinearity started immediately until at ~80V when resistance dropped to $<10\,K\Omega$ and continued to decrease (as seen in Fig. 2). The voltage across the sample was first decreased and then increased. This time resistance was 1,.6 KΩ (@~40V) which continued to decrease at higher voltages. Photo 1 is a representative of the behavior of this sample under continuous-sweep operation mode of the curve-tracer. When high electric powers were delivered to the EC F-563 sample, audible noises and visible arcs were present on the surface of ECF-563. Black streaks was later observed on the surface of the sample indicative of carbonization. The arcing front was originally closer to one of the electrodes and was slowly moving toward the other electrode. High power finally caused the sample to open up. In this communiaction, we will distinguish between the phenomena of switching to 'high current carrying' state (reproducible) and dielectric breakdown-arcing (irreproducible and destructive). The reproducible switching to 'high current carrying' state is the basis for surge arrest material manufacturing industry.

We have observed that the resistance of a virgin ECF-563 preform sample changes widely from sample to sample and it varies with each test. Application of higher currents to EC F-563 continuously increases its conductivity.

Two different behavior thus far have been observed: first, switching to 'high resistive' state in samples of EC F-563 preform sandwiched between gold contact pads [1]; second, switching to 'high current carrying' state in strips of EC F-563. The behavior of the strip samples at very high electric powers can be understood and associated with very high electric field breakdown of insulation and air between neighboring metallic islands. However we are concerned here with reproducible switching to 'high current carrying' state in composite materials. The reported switching in the strip samples is opposite to the earlier observation of switching to 'high resitive' state in EC F-563 samples [1]. One may suspect anisotropy of ECF-563 for this behavior; afterall, some authors [2,3] have seen stratification of silver flakes in the vicinity of surface of some silver filled epoxies. Photo 2 depicts the Secondary Electron Scanning Electron Microscope image of the cross section of ECF-563 preform in the vicinity of its contact with gold plated Kovar plate (the sample is described in [1]) This photograph reveals that silver particles are in variety of shapes and no stratification is observed near the contact interface.

DISCUSSION:

A thin layer of an insulator sandwiched between metal electrodes frequently possess special electrical switching properties which at first can be dismissed as dielectric breakdown. Actually the phenomena of “forming” (reproducible change in electrical conductivity induced by a high electric field) is different from the destructive dielectric breakdown phenomena or arcing.

There are numerous articles in the literature on phenomena of switching in metal-insulator-metal (MIM) junctions (with thin insulators). This subject was mostly considered during 60’s and 70’s and was covered in review articles [4,5]. “Forming” governs the behavior of *as-manufactured* MIM junctions exhibiting switching and which do not require electrical pretreatment (“electroforming”). The current-voltage characteristics of these MIM junctions exhibit S-type or N-type nonlinearity with negative differential resistance (NDR) behavior as depicted in Figure 3. Table 1 is a non-exhaustive list of mechanisms proposed in the literature for N-type or S-type instabilities observed in MIM systems. The most popular theory is carbonaceous filamentation and its rupture indicative of S-type instability. There is no simplified theory which can describe N-type instability. Although some physical evidances for the filament formation are reported in literature e.g; via chemical vapor decoration [1 1], a comprehensive microscopic theory governing both S-type and N-type phenomena in MIM junctions is desired. Both instabilities have been observed in variety of MIM junctions and composites of metal particles in an insulator background; metals vary over a wide range (Ag, Al, Au, Pt, Si, Nb, Be, Mg, Cu, Zn, Ti, Cr, Mn, Fe, Co, Ni, In, Zr, Sn, Pb, Bi, W) and insulators vary from polymers (styrene, acetylene, analine) to oxides (SiO_x, AlO_x, NbO_x, TiO_x, CrO_x, VO_x, TaO_x, CuO_x, MgO) and others (AlN_x,..). What is common among all these systems is metal entities separated by a thin dielectric film. The insulator film is undoubtedly far from an ideal pure dielectric. One can easily envision presence of defects, traps and localized centers in the dielectric. Polymers contain dangling bonds, broken chains, free radicals, spin and charge defects, dopants, *et.cet.*. Oxides fabricated via fast and cheap industrial processes possess well exhibited space charge [12] and are far from single crystalline oxides used in some of MIM junction studies mentioned before. In a typical system, electrons can transfer from one metal entity to the other with variety of mechanisms. These mechanisms involve inelastic interaction of electrons with the defects present in the dielectric material and therefore lead to excess heating, runaway phenomena, and dielectric breakdown.

A microscopic picture of conduction mechanism in thin disordered materials is developed in ref. [1 3]. The authors emphasize the role played by deep localized trap centers in capturing the transit free carriers and the importance of boundary conditions in determining carrier injection and ejection. Reference 13 is our basis for the understanding of switching phenomena observed in ECF-563 as reported in [1]. Space charge may be developed quickly by transit electrons trapped in deep localization centers and may completely block the current flow.

To understand switching to 'high current carrying' state, one needs to distinguish between this reproducible switching phenomena and a destructive mechanism which may finally lead to dielectric breakdown, arcing and carbonization.

A microscopic picture of switching to 'high current carrying' state can be envisioned with the injection of free carriers from metal particles and their free-flight through the thin dielectric material in between. Deep localized traps can capture mobile carriers but it will have minimum effect if the time of flight through the dielectric is much less than the time required by mobile charges to equilibrate with trap centers [14].

In order to tap on a reliable and reproducible switching mechanism for industrial applications, one desires to have electron transfer from one metal entity to the next via quantum mechanical tunneling. Design and fabrication of surge arrest materials with extended life time for industrial applications require knowledge of the true microscopic theory for switching and an exhaustive and broad understanding of dielectrics, metals, and their interfaces.

CONCLUSION:

We have observed switching to both 'high resistive' and 'high current carrying' state in the samples of EC F-563 preform with the gold contacts. Microscopic observations have indicated presence of randomly shaped silver particles embedded in an organic matrix which makes up the bulk of the epoxy. There is also indication that there is a thin layer of epoxy ma rix covering the bulk of silver composite. The picture we have developed describes the injection and ejection of charges between electrodes and the composite and emphasizes the role of deep trap centers capable of capturing free carriers. The space charge developed in between metal entities can stop the current flow as evidenced by switching to 'high resistive' state. Most of fast free-flying carriers injected through the thin dielctric film between metal entities can escape being captured by deep traps and can lead to electrical switching to 'high current carrying' state. Due to the nelastic interaction between transit carriers and defects in the insulator, this high current state will undoubtedly lead to heating and run-away pher omens. A 'high current carrying' state based on quantum tunneling will eliminate heating and can lead to a stable switching with applications as surge arrest materials.

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FIGURE CAPTIONS:

Figure 1- Current-voltage characteristic of a strip sample of ECF-563 (in log-log scale) as measured with 2-probe technique indicates switching to a 'high current carrying' state.

Figure 2- Current-voltage characteristic of a strip sample of ECF-563 as measured with curve-tracer (narrow-pulse operation mode) indicates switching to a 'high current carrying' state.

Photo 1- Current-voltage characteristic of a strip sample of ECF-563 as captured by a camera from a curve-tracer (continuous sweep operation) indicates switching to a 'high current carrying' state.

Photo 2- Secondary Electron Scanning Electron Microscope image of ECF-563 preform and its contact with gold plated Kovar plate has no evidence of stratification.

Figure 3- Drawing depicts S-type and N-type nonlinear current-voltage characteristics.

Table 1- Mechanisms proposed in literature to describe S- and N- type instabilities in current-voltage characteristics of MIM junctions and composite materials.

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Dr. Hamid Javadi is presently at Jet Propulsion Laboratory Spacecraft Communications Technology and Advanced Concepts group. He has three patents on microwave absorption properties of conducting polymers. He is the author of a chapter on Microwave Materials in **The Handbook of Microwave Technology**. The physics of materials such as polymers, dielectrics, high temperature superconductors is his current interest.

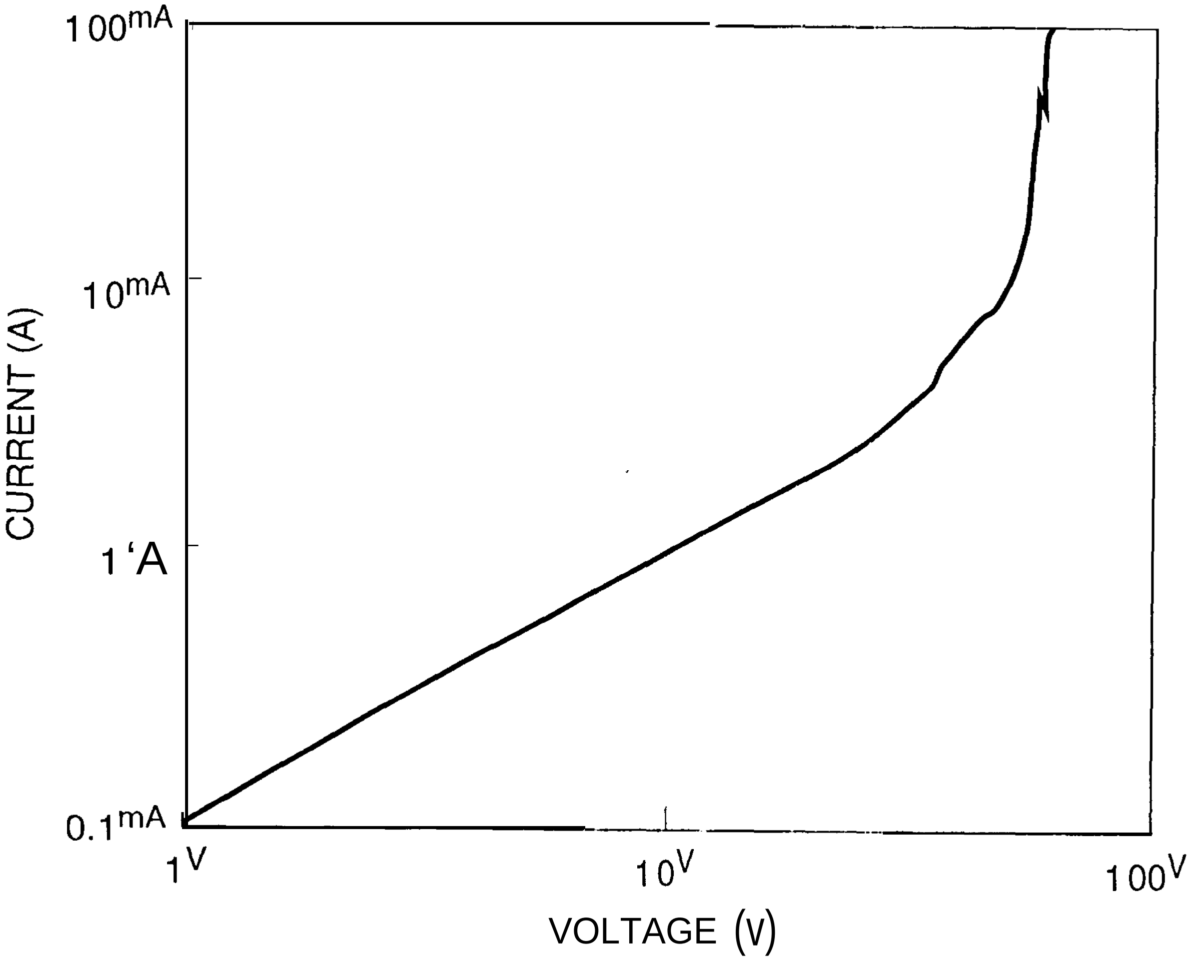


Figure 1 Javadi

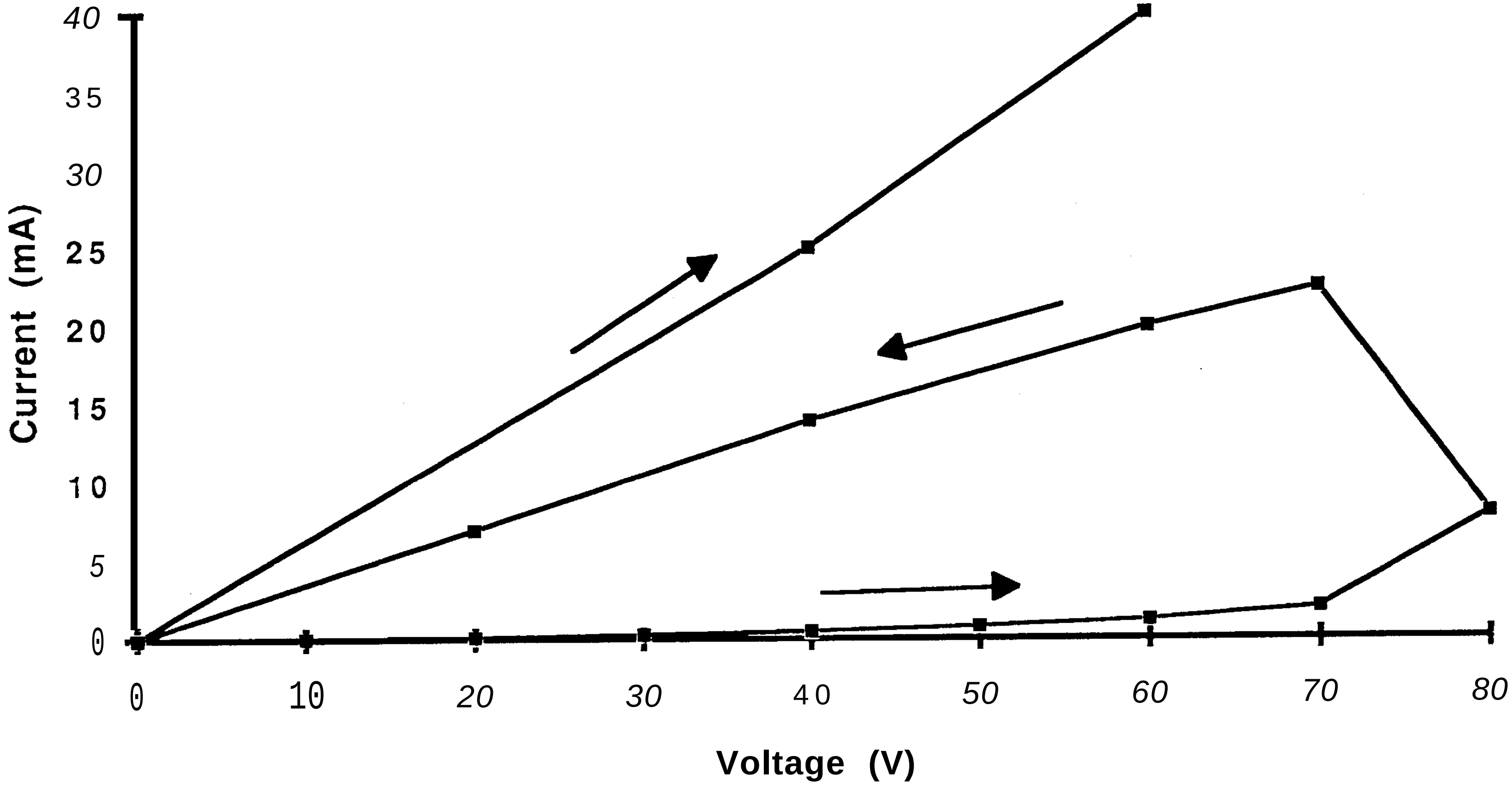


Figure 2 Javadi

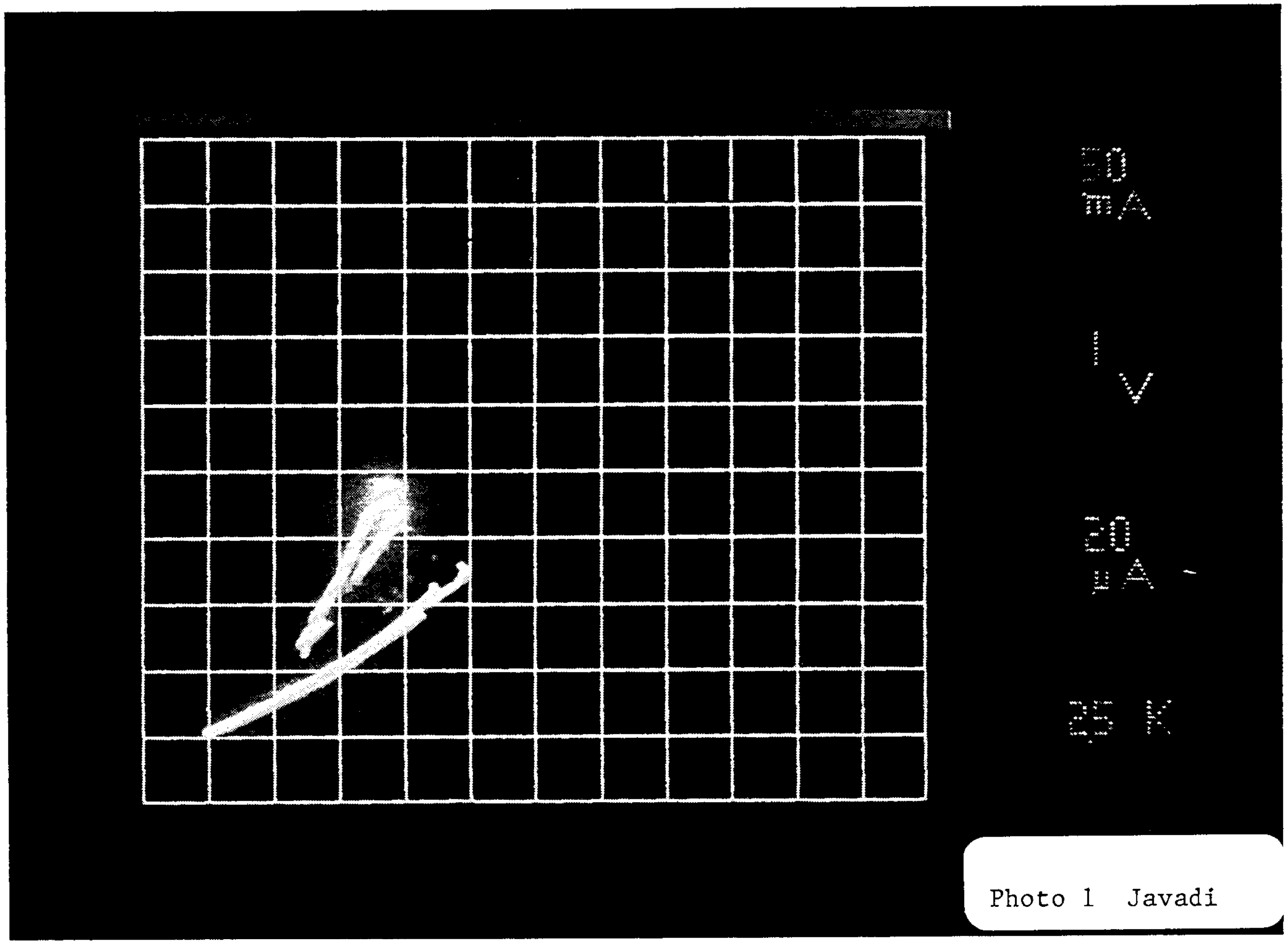
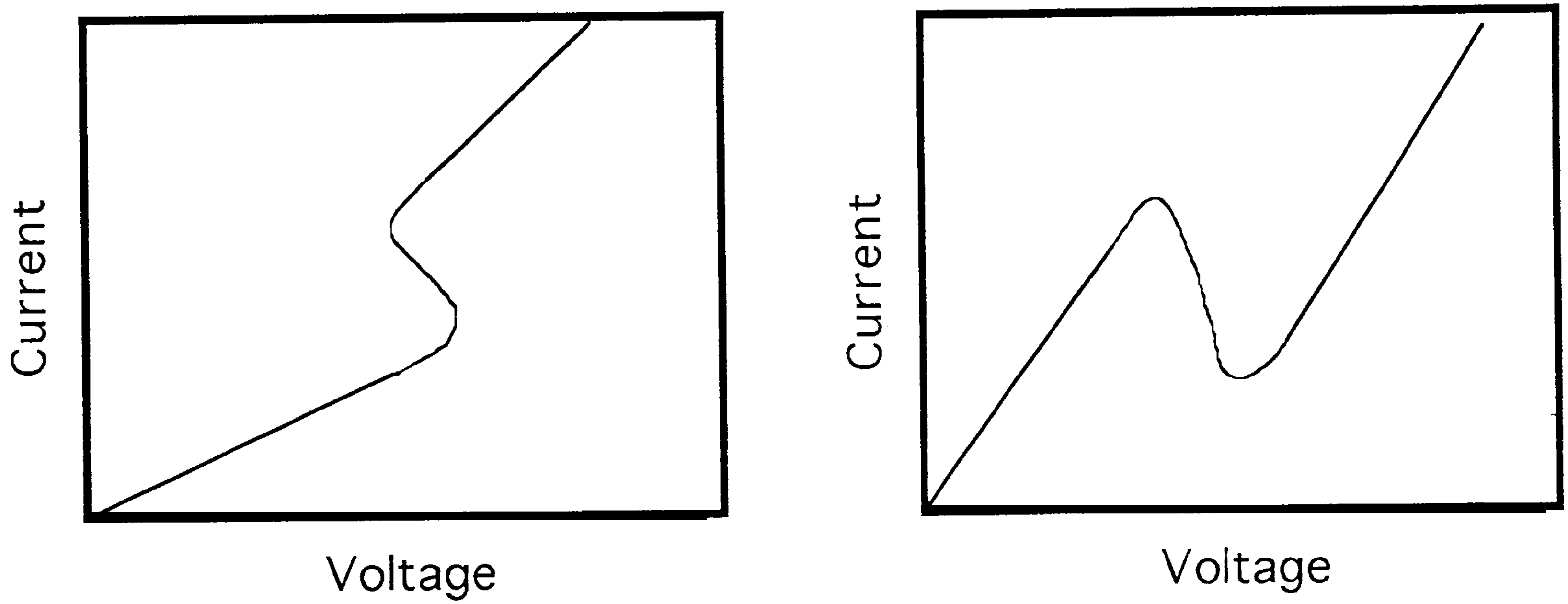


Photo 1 Javadi

Photo 2 Javadi



S-Type N-Type

Figure 3 Javadi

MECHANISM OF NONLINEARITY	TYPE	PHYSICAL SYSTEM	OBSERVATION	REF.
1- ON state: formation of carbon filaments OFF state: filament rupture by Joule heating	S	MIM sandwich		5
2- ON state: formation of carbon filaments with large contact resistance to the metal electrodes. OFF state: filament rupture by Joule heating	N			
3- **ON** state: Formation of metal dendrites OFF state: filament melting by Joule heating	S	Au/NiO/Ni junction		**6**
4- Dielectric breakdown or arcing (creation of plasma)	S			
5- Electron inelastic scattering with charged ions leading to heating	S			
6- Field ionization of traps	S	Al/Al_2O_3-CdS/Al junction	donor density x2 off-on state	7
7- oxidation of metal electrodes in the area of high electric field	N			
8- **ON** state: avalanche injection OFF state: space-charge-limited current	S	$Ta/TaO_2/Au$ junction	bistable switch max. rep. 10^6 Hz	**8**
9- ON state: traps ionized OFF state: traps neutralized	S	$Nb/Nb_2O_5/Pb$ junction		9
10- ON state: filamentary double injection space-charge limited currrent	S	$Ti/TiO_2/Cr$ junction	stable oscillation 1-3 MHz ranae	0

Table 1 Javadi